

**EUR 4615 e**

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**FAST BREEDER FUEL PROCESSING  
PYROCHEMICAL HEAD END STUDY**

by

J.G. WURM, C. DAVID  
(Euratom)

P.R. HEYLEN  
(CEN, Mol)

R.D. DE BEUKELAER, A. DE CONINCK, R. VERBEKE  
(BelgoNucléaire)

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Centre d'Etude de l'Energie Nucléaire - Mol (Belgium)

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Paper presented at  
the European Meeting of Chemical Engineering  
Frankfurt am Main, Germany

June 17-24, 1970



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A general design and lay out of the hybrid Na organic storage pool is given and the inherent advantages over other storage facilities are discussed.

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The presence of Na during the liquid metal (Sb-Cu) dissolution procedure has been investigated and it was shown on hand of small pilot scale experiment that sodium does not present a problem at this stage.

Furthermore, small scale liquid metal dissolution experiments combined with filtration performed on small dummy fuel elements (19 pins) filled with ceramics have proven the feasibility of the process.

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## **ABSTRACT**

The present report describes conceptual head end studies for highly irradiated fast breeder fuel, concerning the storage of short cooled fuels in a hybrid sodium organic coolant storage pool and decladding experiments based on the Sb-Cu liquid metal dissolution technique.

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## **KEYWORDS**

SODIUM  
ORGANIC COOLANT  
STORAGE  
FUELS  
FAST REACTORS

BREEDER REACTORS  
SOLVENT EXTRACTION  
LIQUID METALS  
ANTIMONY  
COPPER

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FAST BREEDER FUEL PROCESSING  
PYROCHEMICAL HEAD END STUDY \*)

INTRODUCTION AND GENERAL CONSIDERATIONS

The investigation of a pyrochemical head end for fast breeder fuel processing is a going programme of S.C.K./C.E.N. Mol which is carried out in close co-operation with BN and Euratom Ispra. While the work performed at Ispra is of a more fundamental nature, the contribution from SCK/CEN and BN concerns mainly conceptual studies, cold pilot experimentation and hot cell laboratory tests.

The fundamental aspects of the pyrochemical head end were presented in several Euratom papers at the ENEA symposium on reprocessing at Mol, October 1969 (1) and at the IAEA panel on reprocessing of highly irradiated fuels at Vienna, May 1969 (2) (3). A conceptual design study including material balances, and a preliminary technical evaluation was given at the "Deutsches Atomforum Reaktortagung", Berlin 1970 (4).

Fig. 1 gives the general flow sheet of the pyrochemical head end as it is conceived for a subsequent aqueous processing. It includes following unit steps :

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\*) Manuscript received on July 27, 1970

- Receiving and handling of the fuel elements at the processing plant
- Storage of the fuel elements
- Liquid metal decanning and fuel separation
- Fuel disaggregation by oxidization either with  $O_2$  or oxidizing molten salts (nitrates)
- Transfer of the powdered fuel to the aqueous dissolver unit
- Fission gas and waste conditioning.

We consider this pyrochemical head end as an alternative to the more classical "chop and leach" which is under development in several countries.

Our flow sheet is based on the following basic options :

- The short cooled F.B. fuel elements are shipped in a container under Na cooling
- The irradiated fuel element handling, storage and decanning are compatible with the presence of Na.

By choosing this route it is aimed to short cut such costly intermediary unit operations, as monitoring of failed fuel elements, Na cleaning, cutting of the ends, disassembling and rebundling of individual pins as it is foreseen for mechanical chopping.

Fig. 2 gives a general view of the hot cell lay-out. To-day we are giving results from the first part of the head end file concerning mainly the fuel storage problem. We are also presenting further laboratory results from the liquid metal dissolution process.



## I. The fuel storage problem

The fuel elements are transferred from the shipping cask under Na cooling directly into a storage pool. Three main problems are predominant for the design of this storage pool :

- The criticality
- The problem of failed or corroded fuel elements and the escape of gaseous fission products
- The heat removal problem of short cooled fuels.

Therefore we have considered the following criteria for the conception of the storage pool :

- Easy heat removal
- Storage consistent with the presence of Na
- No additional cleaning operation
- Limitation of the cell gas contamination
- Minimum capital costs
- Minimum maintenance.

By considering those criteria we have compared different cooling media for an ideal storage pool :

- Water
- Na and NaK
- Inert gas
- Na + organics coolant.

The results of this comparative study have shown that a mixed Na + organics cooling system would satisfy almost all criteria and seems therefore the most suitable one for a F.B. storage pool (5) (6).

Fig. 3 gives the principle of this mixed Na-organic cooling system. Each fuel element is located inside an individual storage tube, filled inside with Na and cooled outside with an organic coolant under forced flow. Each "tube" or "jacket" is closed with a hermetic top lid, and connected to a general gas purification system. At the bottom a Na drain is foreseen for each tube.

Fig. 4 shows the organic cooling loop outside the pool which is composed of a filtration unit, a heat exchanger and a steam preheater. Fig. 5 shows a general view of the pool with individual storage jackets, handling and transfer devices.

TABLE I

Storage pool characteristics

Storage pool	:	Na + organic cooling
Fuel ass. characteristics	:	type Na1
		30 days cooling
		17 kW/fuel ass.
Storage capacity	:	200 fuel ass.
Maximum heat removal	:	3.4 M.W.
Cooling media	:	inside Na
		outside organic (diphenyl)
Flow speed org. (baffles)	:	20 cm/s
Flow capacity org.	:	300 t/h
Inlet temperature org.	:	120 °C
Outlet temperature org.	:	180 °C
Fuel ass. temperature	:	230 °C
Temperature rise on shut-down	:	40 °C/h



### *Advantages of Na-org. cooling*

- No monitoring, cleaning and disassembling
- Efficient and safe cooling
- Reduced capital costs compared to an "all Na" pool
- No fission gas dispersion in the cell atmosphere
- Storage pool can be under air atmosphere
- Reduced labour costs
- High safety
- Existing and improved technology for the organic media.

### *Organic coolant behaviour*

Since the outlet temperature of the organic coolant is kept at only 180 °C, pyrolysis under normal operating conditions is negligible. However, in case of a shut down of the pumping system, the temperature rise of the fuel element would only be 40 °C/h which is an acceptable value.

Concerning the radiolysis of the organic coolant it should be emphasized that the  $\gamma$ -rays are strongly attenuated by the geometrical configuration of Na-filled storage jacket. Calculations made by Van Bosstraeten S.C.K./C.E.N. Mol have shown that only about 10 % of the  $\gamma$ -rays will affect the organic coolant, so that only 0.1 % of the organic coolant should be replaced per year (7).

## II. Liquid metal decladding

### II.1. Na dissolution in Sb-Cu solvent alloy

According to ORNL (8) the amount of Na attached to a fuel assembly is estimated to be 3 kg. It was necessary to investigate that this quantity could be dissolved and retained in the solvent alloy at the operating temperature of 1000 °C (1 wt.% Na in Sb-Cu 10 wt.%).

The Sb-Na phase diagramme shows that Na will be dissolved in any quantity in Sb. Also the existence of  $\text{Sb-Na}_3$  and Na-Sb intermetallic compounds indicate a strong affinity between Sb and Na (Fig. 6). No phase diagrammes for Na-Cu, Na-Fe, Na-Cr and Na-Ni are available. The affinity of Na for these metals will therefore be neglected.

EMF orientation measurements of the Na/Sb - Na (1 %) couple have been performed. From 0.7 V at 670 °C an activity coefficient of  $3 \times 10^{-2}$  was derived. At 1000 °C this activity coefficient can be extrapolated to the value of  $7 \times 10^{-2}$ . Since the vapor pressure of Na over liquid Na at 1000 °C amounts to about 2.5 atm, it is possible to calculate that the vapor pressure of Na over Sb-Cu 10 wt.% - Na 1 wt.% alloy at 1000 °C will be as low as 1 mm of Hg.

The Na volatility out of a Sb-Na 1.3 wt.% alloy and a Sb-Na 0.8 wt.% - stainless steel 20 wt.% alloy was measured at Euratom Ispra by M. Payrissat.



*Operating conditions*

The alloy was heated in a graphite crucible. Samples of the Sb-Na alloy were taken periodically by means of a capillary quartz tube.

TABLE II  
Typical Na concentration in solvent alloy

	Temperature °C	Sampling time min	Na concentration wt.%
Run No. 4 Sample Sb-Na (1.3 wt.%)	800	30	1.28
		60	1.32
		90	1.38
		120	1.4
		150	1.32
Run No. 5 Sample Sb-Na (0.8 wt.%) + SS 20 wt.%	900	20	0.7
		40	0.76
		60	0.76
		80	0.74
		100	0.72
		120	0.74

These results indicate that the Na will be dissolved and remains trapped in a Sb-Cu 10 wt.% solvent alloy at the decanning operating conditions.

## II.2. Liquid metal decanning laboratory experiments

The dissolution kinetic of stainless steel in the Sb-Cu solvent alloy, reported previously (1) (2) (3) has shown that the dissolution rate is strongly depending on the temperature and the diffusion of the stainless steel in the liquid solvent alloy. To remove the stainless steel saturated solvent alloy layer surrounding the solid stainless steel agitation was necessary. The simplest and most adequate agitating technique seems to be the hydraulic pulsation of the solvent alloy inside the fuel assembly in order to dissolve the fuel pins more rapidly than the wrapper. To prove the feasibility of this dissolution technique under the most real conditions, a series of laboratory experiments on two types of dummy fuel assemblies have been performed.

### *Type No. 1*

Two empty fuel pins ( $\emptyset$  10 mm, thickness 0.3 mm, stainless steel 340 L) are fixed inside a wrapper tube ( $\emptyset$  25 mm, thickness 2.5 mm, length 300 mm) with a connection for the pulsating device.

### *Type No. 2*

19 fuel pins ( $\emptyset$  10 mm, stainless steel 304 L) filled with  $ZrO_2$  pellets and powder are fixed inside a hexagonal wrapper tube (56 mm and 300 mm length) connected to the pulsating device.

The laboratory set-up is given in Fig. 7. The dummy fuel element is placed inside a graphite cylinder equipped with a fuel filtration device at the bottom and connected to a piston pump at the top.

The solvent alloy can be filtered, either under vacuum or pressure. A T.C. located also in the graphite is connected to a temperature recorder. The whole set-up is placed inside a quartz tube vacuum furnace and heated by induction (10 kHz, 15 kW). The modulation amplitude is varied by the variable piston course. A medium pulsation cycle lasts for about 10 s.

#### *Performance of fuel type No. 1*

The results from the first experimental series indicate that the differential dissolution of the pins and the wrapper with hydraulic pulsation is strongly dependent on the temperature and the stainless steel content in the solvent alloy.

##### *- Dissolution temperature 750 °C*

With fresh solvent alloy, the pins are completely dissolved, and the wrapper partially, in about 20 min of pulsation.

##### *- Dissolution temperature 1000 °C*

With fresh solvent alloy the same result is obtained in 4 min. With a solvent alloy containing already more than 15 wt.% of stainless steel the dissolution of the pins is completed in 5 min with only a limited attack of the wrapper. Further tests are still necessary to optimize the dissolution temperature as a function of the stainless steel content.

*Performance of fuel type No. 2*

With a fresh alloy at 1000 °C a complete dissolution of the wrapper (1 mm thickness) and almost all the pins were dissolved in about 2 min. Since the dissolution speed was too fast, the experiments will be repeated at lower temperatures (900, 850 and 800 °C).

II.3. Filtration

The fuel solvent alloy filtration parameters were investigated in a separate set-up Fig. 8. The filtration apparatus is composed of a HF heated graphite cylinder equipped with a porous graphite filter at the bottom and a rotating agitator, the solvent alloy can be forced through the filter either by vacuum or pressure or both.

Filter characteristics : Union Carbide, grade 60  
Diameter : 24 mm  
Thickness : 10 mm  
Porosity : 48 %  
Mean pore diameter : 0.043 mm

TABLE III  
Filtration results

	I	II
Temperature	1000 °C	1000 °C
ΔP over filter in mm Hg	330	950
Filtration time in min	1	10
Solvent alloy Sb-Cu 10 wt.% + S.S. 30 wt.%	180 g	180 g
Fuel UO <sub>2</sub> grain size < 80 μ	0	18 h



The separation of the fuel and the solvent alloy was complete and no fuel losses could be detected in the filtered solvent alloy. However with powdered fuel the differential filtering pressure must be increased.

### CONCLUSIONS

The above described laboratory experiments (Ispra) will be completed in about 3-4 months. We are already preparing cold pilot experiments on a representative scale with F.B. dummy fuel elements (1 m length) in order to demonstrate the feasibility of an integrated liquid metal dissolution and filtration unit.

In parallel the laboratory work on decanned fuel with molten salts concerning mainly the disaggregation and the U-Pu separation will be continued on a laboratory cold pilot scale.

Finally a head end engineering file based on a technico economical evaluation is under study and will be issued at the end of 1970.

### ACKNOWLEDGEMENTS

The authors are grateful to M. Payrissat, Euratom Ispra, for his contribution to Na solubility measurements. They express their gratitude to A. Hoffmann, Euratom Ispra, for his skilful participation in the laboratory experiments and to R. Botte, S.A. BELGONUCLEAIRE Mol, for his valuable conceptual design.

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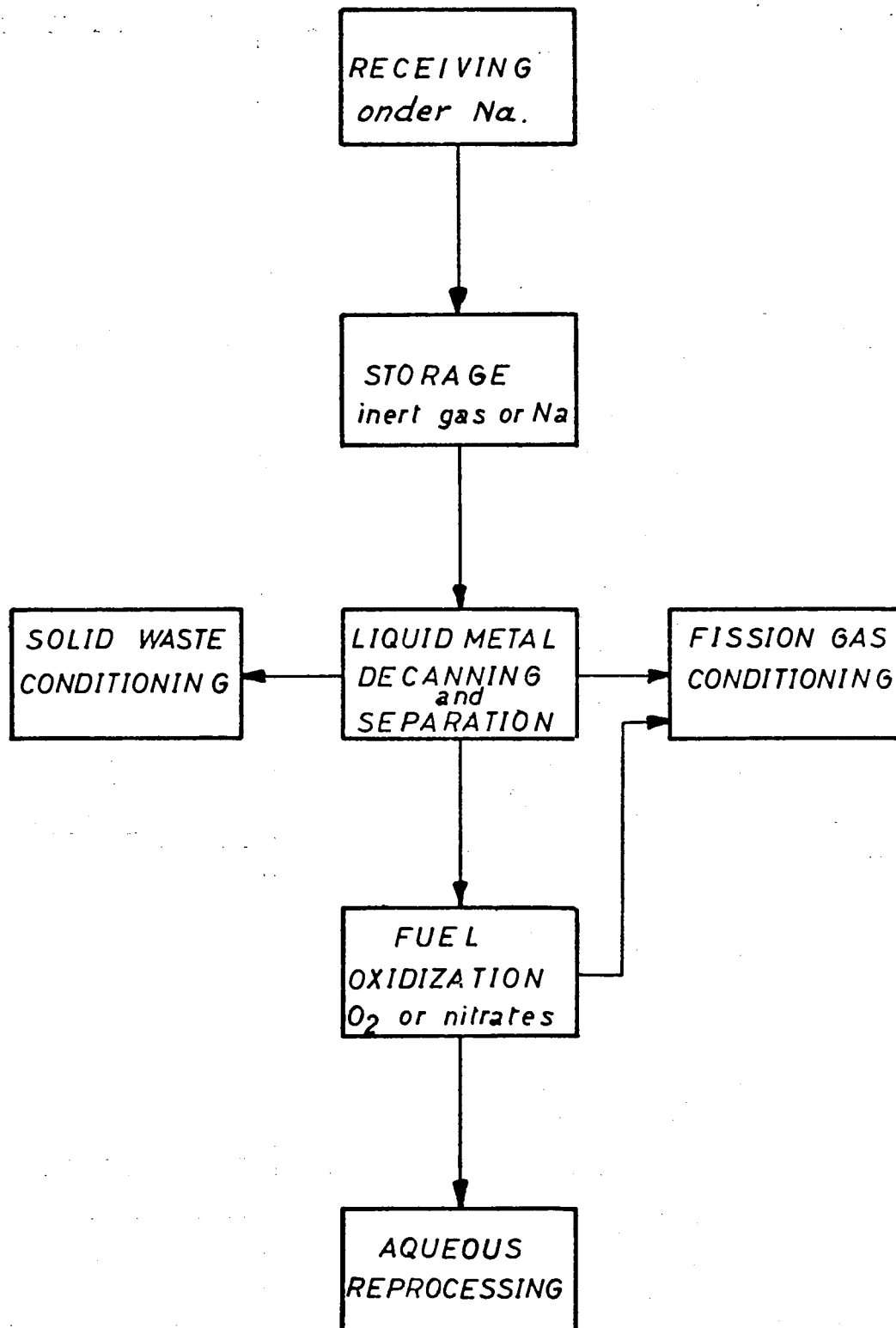
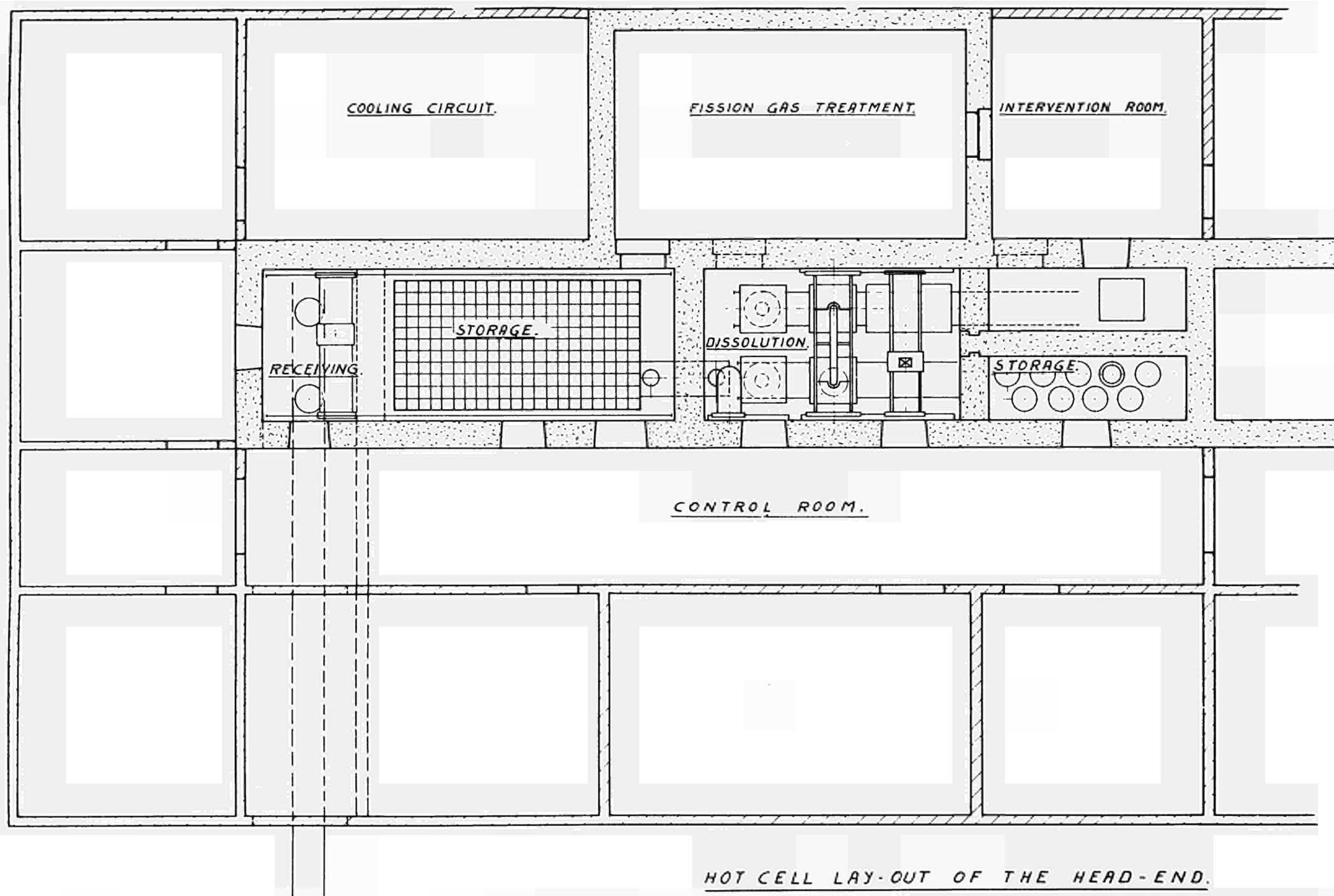


FIG:1

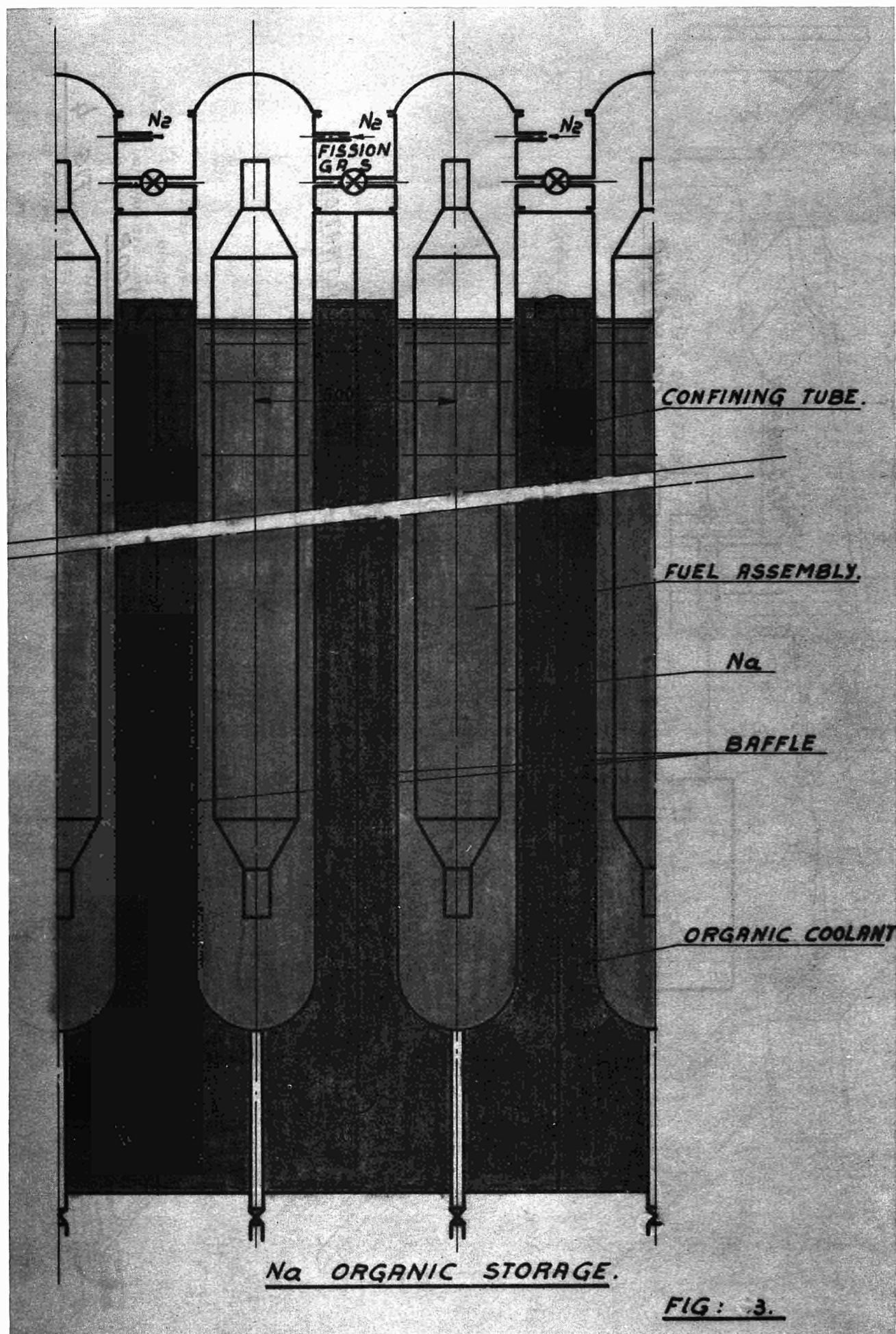
INTEGRATED HEAD END FLOW SHEET

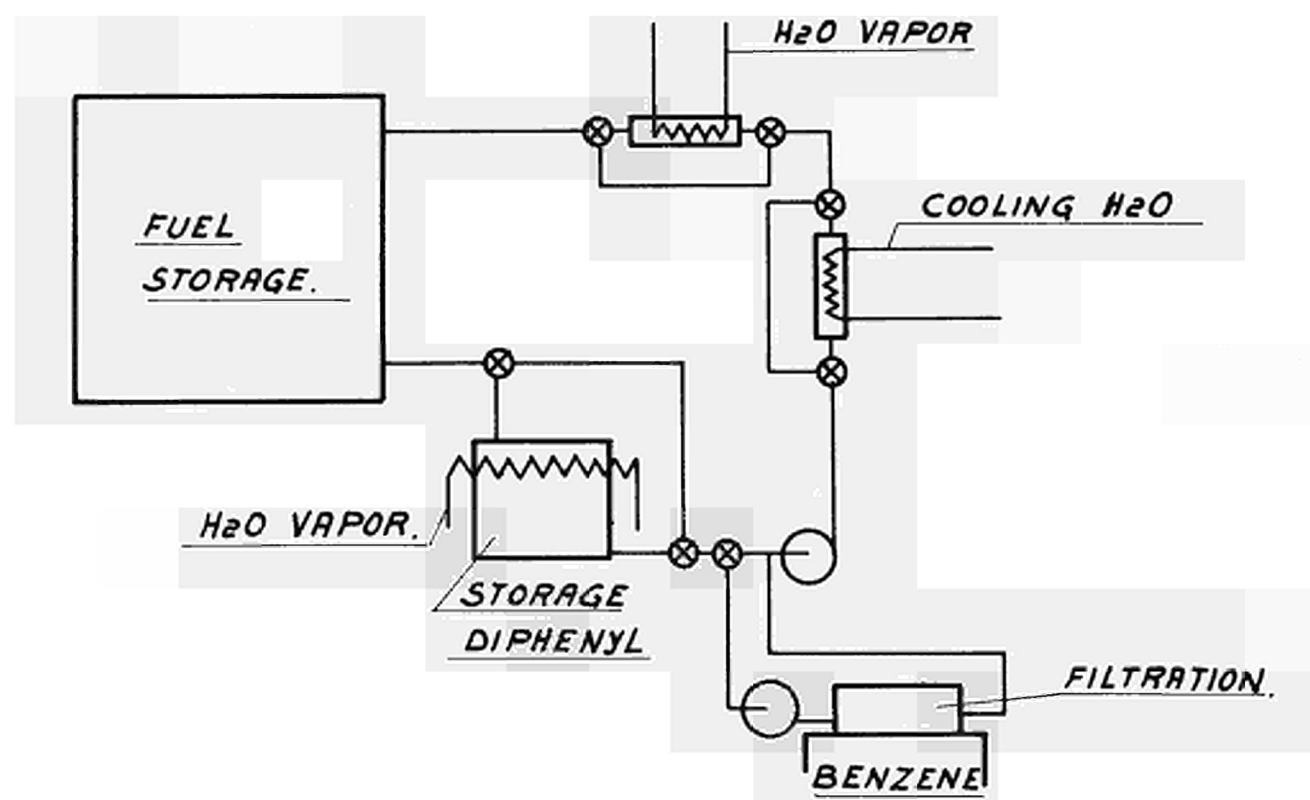


HOT CELL LAY-OUT OF THE HEAD-END.

FIG. 2.



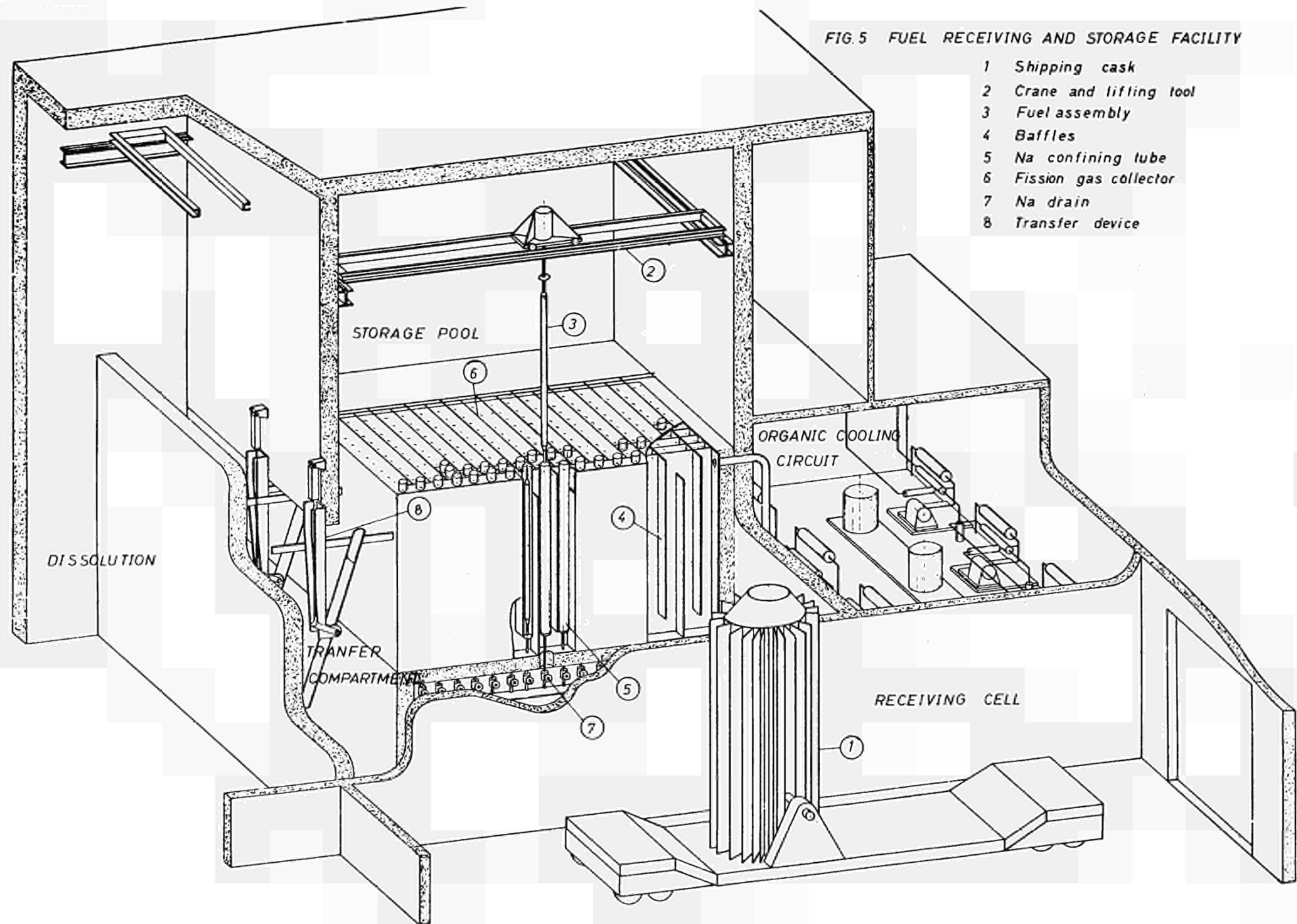


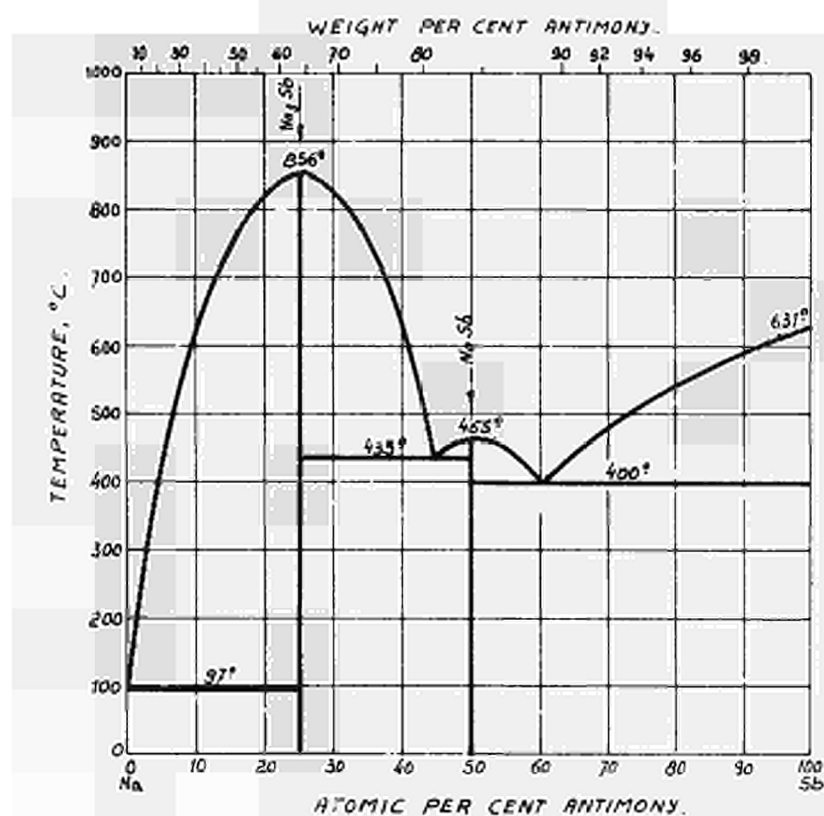


GENERAL ORGANIC COOLING LOOP.

FIG: 4.

FIG. 5 FUEL RECEIVING AND STORAGE FACILITY

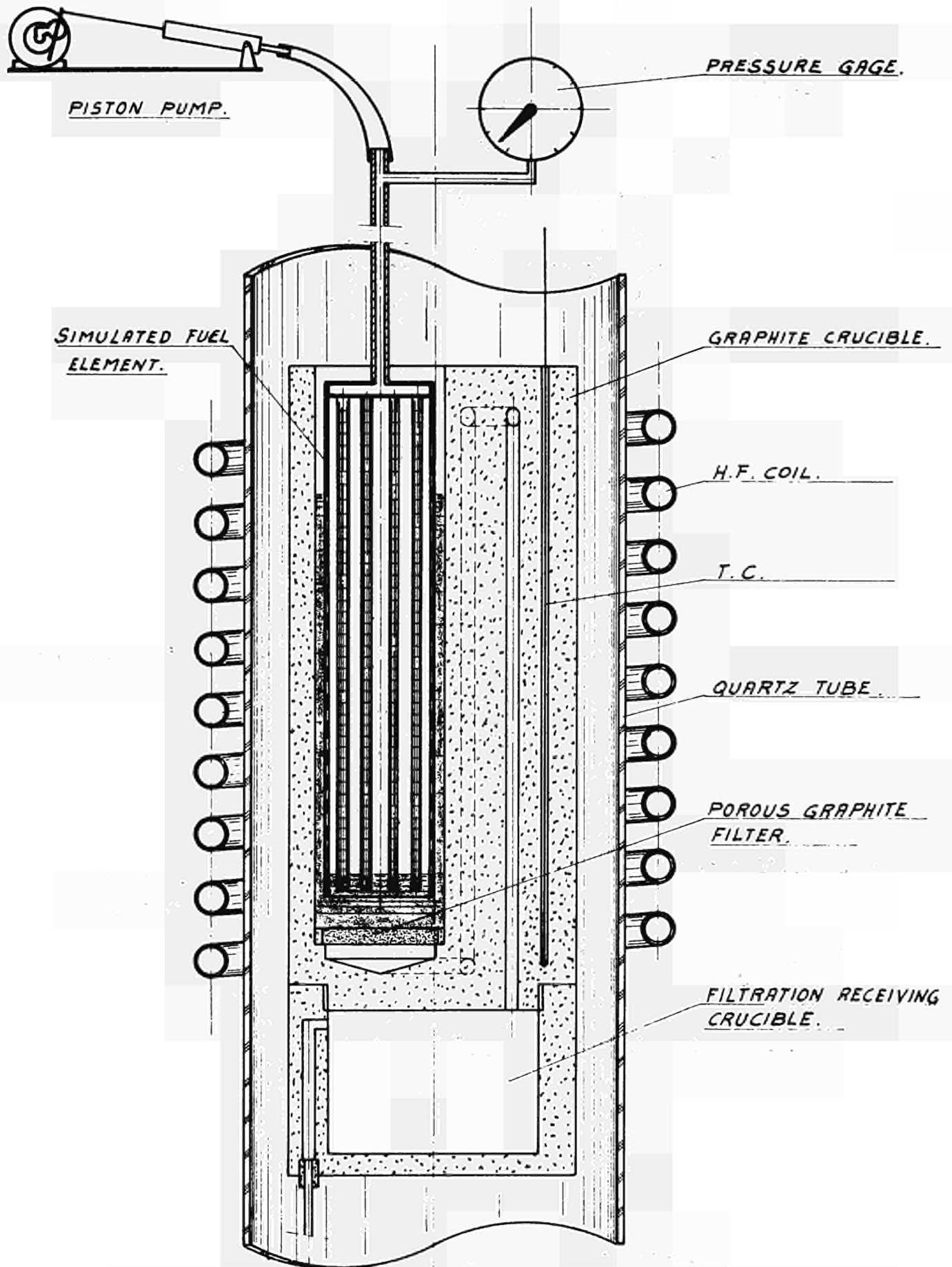




PHASE DIAGRAM Sb - Na.

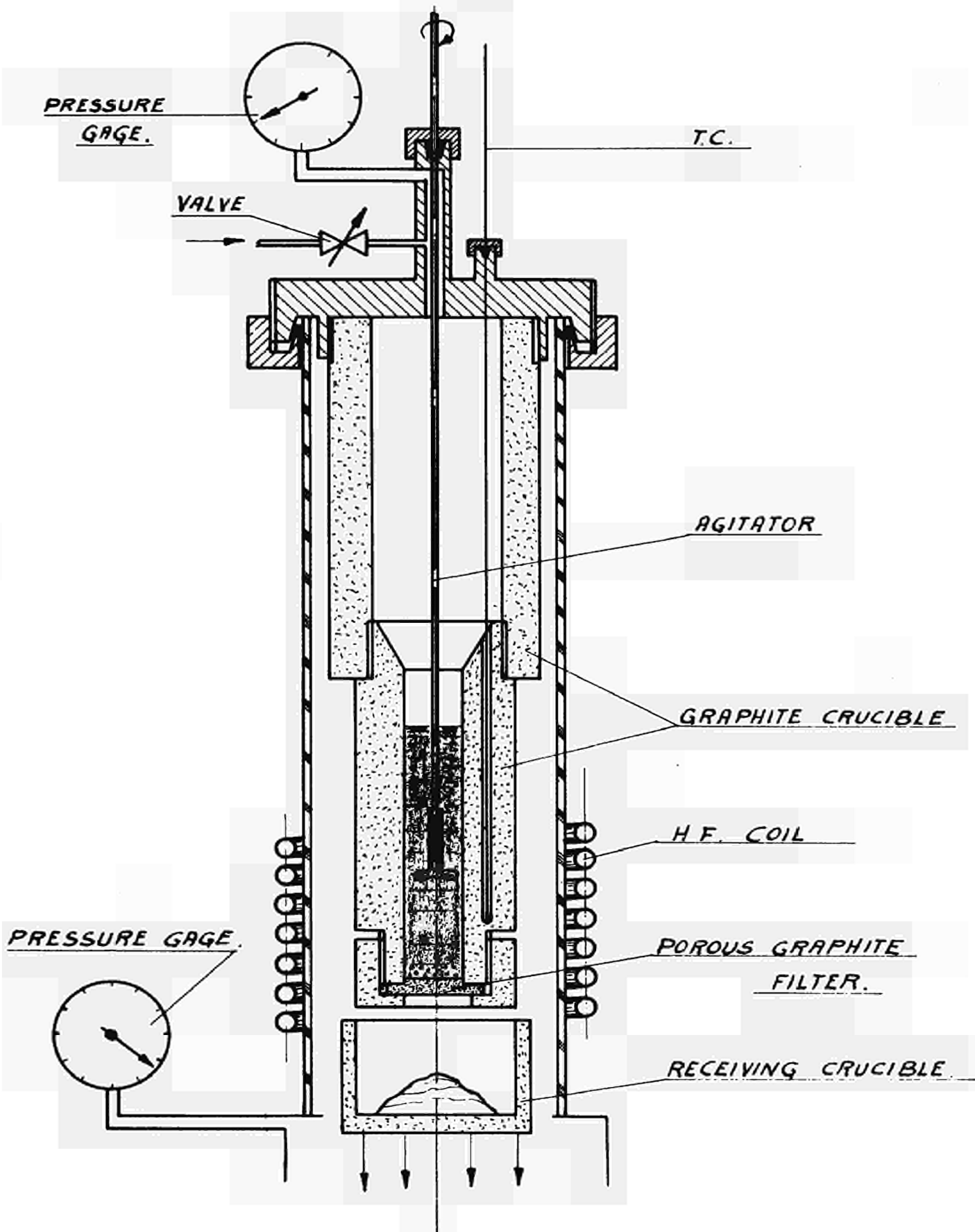
FIG: 6.





PULSATING DISSOLUTION TECHNIC ON REDUCED FUEL ELEMENT.

Fig: 7.



LABORATORY FILTER UNIT.

FIG : 8.



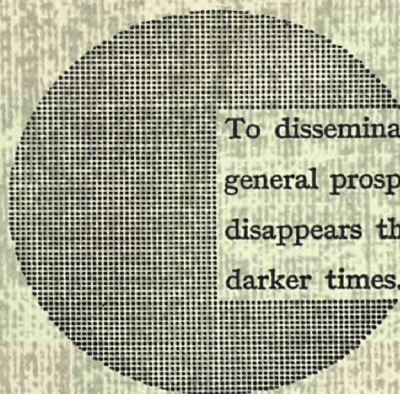
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To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

**Alfred Nobel**



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